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In the manuscript originally published online, there were minor errors in Figure 3C. They have been corrected here.

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A Quantitative Synthesis of Mercury in Commercial Seafood and Implications for Exposure in the U.S.

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Abbreviations:

EPA: U.S. Environmental Protection Agency

FDA-MP: U.S. Food and Drug Administration Hg Monitoring Program

Hg: Mercury

MeHg: Methylmercury

NMFS: National Marine Fisheries Service

Abstract

Background: Mercury (Hg) is a toxic metal that presents public health risks through fish consumption. A major source of uncertainty in evaluating harmful exposure is inadequate knowledge of Hg concentrations in commercially important seafood.

Objectives: We examine patterns, variability and knowledge gaps of Hg in common commercial seafood items in the U.S. and compare seafood Hg concentrations from our database to those used for exposure estimates and consumption advice.

Methods: We developed a database of Hg concentrations in fish and shellfish common to the U.S. market by aggregating available data from government monitoring programs and the scientific literature. We calculated a grand mean for individual seafood items, based on reported means from individual studies, weighted by sample size. We compared database results to those of federal programs and human health criteria (U.S. Food and Drug Administration Hg Monitoring Program, FDA-MP, U.S. Environmental Protection Agency, EPA).

Results: Mean Hg concentrations for each seafood item were highly variable among studies, spanning 0.3 to 2.4 orders of magnitude. Farmed fish generally had lower grand mean Hg concentrations than their wild counterparts, with wild seafood having 2 to12-fold higher concentrations, depending on the seafood item. However, farmed fish are relatively understudied, as are specific seafood items and seafood imports from Asia and South America. Finally, we found large discrepancies between mean Hg concentrations estimated from our database and FDA-MP estimates for most seafood items examined.

Conclusions: The high variability in Hg in common seafood items has considerable ramifications for public health and the formulation of consumption guidelines. Exposure and risk analyses derived from smaller datasets do not reflect our collective, available information on seafood Hg concentrations.

Introduction

Human exposure to Hg from seafood consumption and attendant risks are difficult to estimate and are often the subject of intense debate. However there is broad recognition of the need for large-scale information on mercury (Hg) concentrations in marine fish and shellfish in order to better understand and control Hg exposure and risk (National Research Council 2000). While U.S. seafood consumption has plateaued in recent years, global seafood demand is on the rise (Food and Agriculture Organization of the United Nations 2010; National Marine Fisheries Service 2011a). Fish and shellfish are important sources of lean protein and other nutrients, including essential omega-3 fatty acids, which confer important health benefits (Albert et al. 2002; Huynh and Kitts 2009; Simopoulos 1991; Siscovick et al. 1995). However, all seafood also contains mercury, primarily in the form of methylmercury (MeHg). In sufficient doses, MeHg can cause adverse neurodevelopmental (Myers et al. 2009; Oken et al. 2005; Trasande et al. 2005), cardiovascular (Grandjean et al. 2004) and immunological health effects (Gardner et al. 2010). Since the majority of human exposure to MeHg is through seafood consumption (IPSC 1990, 1991; National Research Council 2000; UNEP (United Nations Environment Programme) 2003), it is critical to have reliable estimates of Hg concentrations in seafood items in order to confidently identify those that are low in Hg. Such efforts will better inform estimates of exposure and risk and help consumers make decisions about the types and quantities of seafood that are both safe to eat and nutritionally beneficial.

Seafood Hg concentrations can be highly variable, even within the same species (National Research Council 2000; Sunderland 2007). While hundreds of individual studies collectively have monitored fish mercury concentrations around the world, we still have an incomplete understanding of general Hg patterns, particularly in commercial fish and shellfish

from marine waters (Chen et al. 2008). Moreover, our knowledge of the extent of Hg variability is limited. Aggregating data from individual studies is necessary to obtain a clearer understanding of general patterns in Hg content of commercial fish. To date, the largest, most well-known existing databases on Hg content in U.S. commercial fish were developed by federal government agencies (National Marine Fisheries Service (NMFS), FDA). However, the NMFS study from the 1970s (Hall et al. 1978) is relatively outdated and the FDA-MP (FDA Monitoring Program 2011; USFDA 2011) contains smaller sample sizes and fewer species. In contrast, data from intensive, small-scale studies that focus on obtaining large sample sizes of a specific taxon are less susceptible to random sampling error and are likely to yield better estimates of central tendency. Such smaller, intensive studies are common within the scientific literature (e.g. Adams and McMichael 2007; Burger and Gochfeld 2006), but typically are not integrated into larger analyses of exposure and risk. Finally, federal databases, particularly the NMFS study, may not accurately reflect Hg concentrations of imported fish, even though the amount of imported, edible seafood consumed in the U.S. is increasing (National Marine Fisheries Service 2010). Imports now account for more than 80% of the seafood eaten in the US (National Marine Fisheries Service 2011a). Thus, the inclusion of Hg data for imported seafood would fill a crucial knowledge gap. Combining data from government and academic sources would allow for more precise estimates of mercury concentrations in U.S. imported and domestic seafood items using the broadest available knowledge base.

We examined patterns of Hg concentrations in U.S. commercial seafood items using, what is to our knowledge, the largest compilation of available academic and agency data to date. Our overarching goal was to examine longstanding questions about the patterns of seafood Hg concentrations and their variability. Our Seafood Hg Database aggregates Hg measurements of

hundreds of seafood items from federal and state agencies, and smaller, more intensive studies from the scientific literature. Our specific goals were to reliably identify low Hg and high Hg fish, and to identify the seafood items and geographic regions that are in most need of further study. Additionally, we compared Hg concentrations in farmed seafood items relative to concentrations in their wild seafood counterparts. Finally, we compared Hg concentrations for individual seafood items to those summarized from the FDA-MP (FDA Monitoring Program 2011). The FDA-MP data are commonly used for risk assessment and exposure estimates (Ginsberg and Toal 2009; Sunderland 2007; Tran et al. 2004), and in the development of state-level consumption advice for consumers (e.g. State of Maryland, 2011; State of Minnesota, 2011). At least one previous study has compared FDA-MP data to those from independent studies in order to better estimate Hg intake in the U.S. (Sunderland 2007). Our study builds on this approach by synthesizing a much larger aggregation of available data in order to better characterize Hg variability and assess the current state of knowledge of seafood Hg content. Ideally, these improved estimates of Hg concentrations in commercial fish will help enable more accurate assessments of potential exposure and inform both public health programs and the public itself regarding the types and amounts of fish that are safe to eat.

Methods

Data Gathering and Inclusion Criteria

Our guiding principle for building the Seafood Hg Database was to focus on fish and shellfish from sources that could reasonably be sold in the U.S. Our database was developed to reflect the range of possible Hg levels for seafood items thought to be the top contributors to human Hg exposure in the U.S. because they are relatively high in Hg and/or they constitute

relatively large shares of the US seafood market (top 51 Hg contributors defined in Groth 2010). Detailed taxonomic and geographic harvest information is often lacking or incorrect in the seafood marketplace (Jacquet and Pauly 2008). Thus, our database does not model the exact composition of the US seafood market. Rather it reflects the range of seafood species and seafood Hg concentrations that are available to U.S. seafood consumers.

Data were gathered from federal and state government reports and from the scientific peer-reviewed literature. We obtained data from federal and state government agencies that either made their fish tissue monitoring results publically available online (e.g. USFDA raw data (USFDA 2011), USEPA, State of Virginia, State of North Carolina, State of New Jersey) or provided data upon request (e.g., State of Delaware, State of Hawaii). In addition, we searched for published, peer-reviewed papers indexed in Web of Science before December 15, 2010. We conducted literature searches for individual seafood items based on seafood varieties listed as the top 51 Hg contributors to the U.S. population (Groth 2010). Search terms included "mercury" and the common names of these fish or shellfish (e.g. "mercury and salmon") (see Supplemental Material, Search Terms for Table S2).

From the data gathering and search results, we included fish and shellfish from sources that were likely to enter the US seafood market. We included data on edible portions (fillet or whole fish) of any fish or shellfish species likely to be included in the top 51 seafood varieties (e.g. "redfish" were included with "ocean perch") based on federal commercial fisheries landings (fisheries landed and sold in the U.S.) and seafood import statistics (National Marine Fisheries Service 2005). Mercury concentrations in whole fish can be lower than concentrations in fillets (Goldstein et al. 1996), probably because mercury is primarily associated with muscle tissue. Thus, the inclusion of data based on fillets as well as whole fish, which are common in the

market particularly for smaller fish such as anchovies, may underestimate fish Hg content relative to those based on fillet only. We classified seafood items as being from domestic or imported sources based on geographic locations specified in the original study. We assumed that all marine fish caught commercially from domestic waters were relevant to the US market. Data for a given fish or shellfish species collected from market basket studies or direct harvest from countries outside of the US were only included if at least 5% of all imports of that species into the US fisheries market (by volume) were from that country according to NMFS import statistics as of 2010 (National Marine Fisheries Service 2011b). In addition, imported seafood items that did not meet this criterion were included if the samples were collected from water bodies connected to other countries that meet this criterion. Highly migratory fish caught from major ocean basins, (tuna, shark and swordfish) were included regardless of country of origin.

Of the top 51 seafood varieties, less than 10 are freshwater fish. For most freshwater items collected from domestic waters, we included data from the Great Lakes, because the Great Lakes are the main sources of these species to the market (National Marine Fisheries Service 2011c). We did not include salmon species from the Great Lakes, because the commercial catch of salmon from the Great Lakes has been negligible for at least one decade (Baldwin et al. 2009). For striped bass (*Morone saxatilis*), we included data for wild fish only from Atlantic states, because commercial fisheries do not exist for this species in the Gulf of Mexico or Pacific coast (National Marine Fisheries Service 2007). For catfish, carp and perch, we included fish collected from Atlantic or Gulf coast states that report commercial landings of these fish (National Marine Fisheries Service 2007), excluding samples from interior or landlocked freshwater sources. Data for farmed species of commercial freshwater fish were included if the fish were specifically

raised for consumption (e.g. farmed catfish), and the fish were of market size (versus juvenile fish from hatcheries) and were fed conventional feed (e.g. Berntssen et al. 2010).

Exclusion Criteria

We screened approximately 1000 government monitoring programs and peer-reviewed academic studies for inclusion. Upon critically examining each study, we excluded entire datasets, or select data from studies, based on one or more of the following criteria:

- Data resulting from experimental Hg exposures.
- Data on fish or shellfish that are not a primary source of commercial fish to US consumers, based on the geographic location of collection.
- Studies that were not written in the English language.
- Data that were repeated from another source already included the database. Examples include data repeated in review papers as well as original papers, or data repeated in aggregate federal government databases (e.g., EPA National Listing of Fish Advisories) and original state data sources (e.g., State of North Carolina). Duplicate entries were routinely screened for and excluded from all calculations.
- Data for fish from locations with known point source Hg contamination or associated fisheries closures.
- Data for young-of-year fish (born within the past year). However, we included Hg values from other, smaller body-size fish which may be excluded from the US market due to catch restrictions. Hg concentrations tend to be lower in small fish compared to larger fish of the same species, thus may underestimate true average of Hg values in US commercial fish.

- Studies conducted by non-governmental organizations, public interest groups or news media that were not peer-reviewed or incorporated into government monitoring efforts.
- Studies that did not report the necessary Hg data (raw data, or arithmetic mean Hg or MeHg concentration and sample size). For example we excluded studies that presented Hg concentrations in a graph, or as a range, geometric mean, or median. Geometric means and medians were rarely reported in the literature. Therefore, we only included arithmetic mean Hg concentrations, or calculated arithmetic means based on raw data when reported.
- Data from areas with no commercial fishing activity, such as no-take marine reserves and national parks (e.g., Rencz et al. 2003; Wyn et al. 2009).

Data Extraction

We extracted mean Hg concentrations (ppm, wet weight), sample size and geographic location for each seafood item reported in each study. Approximately 40% of the included sources reported standard deviations, or standard errors. Thus, analyses requiring standard deviations would exclude the bulk of the dataset. Therefore, we focused on examining mean Hg concentrations in the interest of including the range of Hg concentrations for each seafood item using the largest possible dataset. We extracted total mercury values whenever possible, but used methylmercury values when these were reported instead of total mercury. Approximately 95% of total Hg in fish muscle tissue occurs in the form of methylmercury (Bloom 1992). Therefore, we assumed that methylmercury concentrations are similar to total Hg concentrations. Nevertheless, because methylmercury concentrations are lower than total Hg, our calculated, grand mean mercury concentrations for certain seafood items may be slightly lower than if based solely on total Hg concentrations. Mercury values reported as dry weight concentrations were converted to

wet weight concentrations according to moisture content if reported, or assuming 80% water content. When Hg concentrations were reported as non-detect (approximately < 10% of all database entries), values were entered as one-half the detection limit from the study (Clarke 1998), or were excluded when detection limits were not reported.

When a study reported multiple mean Hg values for a given seafood item (Hall et al. 1978), we calculated a weighted mean, using sample size for the mean as the weight. When a study reported multiple Hg values for a given seafood item, but did not provide sample sizes for individual values (e.g., Cossa et al. 1992; Deshpande et al. 2009; Jackson 1991), we assumed sample sizes were equivalent across values. Thus, overall means calculated from these studies were not weighted.

Data Analysis

We calculated an aggregate, grand weighted mean ($\overline{Hg}_W$) for each seafood item based on means weighted by sample size across studies

$$\overline{Hg}_w = \frac{\sum(Hg_i \times w_i)}{\sum w_i} \qquad [1]$$

where Hg_i is the ith reported mean and w_i is the weight (reported sample size) of the ith observation. We estimated variability of Hg in seafood items by calculating a weighted, grand standard deviation, corresponding to the grand mean. The Seafood Hg Database comprises mean mercury values reported by individual studies as observations, as opposed to raw Hg data values. By definition, the standard deviation of sample means is the standard error of the global distribution of Hg values. Therefore we estimated the weighted standard error (SE_w) of the distribution underlying the grand mean using the formula for the weighted standard deviation,

$$SE_w = \sqrt{\frac{\sum_{i=1}^{N} w_i (Hg_i - \overline{Hg}_w)^2}{\frac{(N-1)\sum_{i=1}^{N} w_i}{N}}} \qquad [2]$$

where N is the number of studies from which mean Hg values were collected. To obtain the weighted standard deviation (SD_w) of the global distribution, we multiplied SE_w by the square root of average sample size across studies for each seafood item, yielding the formula

$$SD_w = \sqrt{\frac{\sum_{i=1}^{N} w_i (Hg_i - \overline{Hg}_w)^2}{(N-1)}} \qquad [3]$$

Monte Carlo simulations tested for potential bias of Eq.3 using hypothetical data approximating the composition of the database. Specifically, we simulated a true standard deviation of the global distribution using random numbers drawn from normal and lognormal distributions, where w_i ranged from 2-100 and N ranged from 50-300. Tests of 10,000 replicates demonstrated that Eq. 3 was an unbiased estimator of the true standard deviation of the global distribution, and was insensitive to the type of distribution used and variation in sample size (data not shown).

We calculated the grand mean, grand standard deviation, range (minimum and maximum reported mean), coefficient of variation, and total number of samples across all studies for the seafood item names searched (e.g., salmon), as well as for seafood items with higher taxonomic resolution within the search results (e.g., Atlantic salmon) and broader taxonomic categories for specific analyses. Thus, results are presented for a larger number of seafood groups than the original top 51 seafood items from the search. We compared our findings to summarized Hg data of the FDA-MP accessed September 15 2011 (FDA Monitoring Program 2011) for seafood items for which direct comparisons were possible given available data (58 seafood items). In some cases, seafood items were grouped together into larger seafood categories, which often included

multiple taxa. For example, for direct comparison with Hg concentrations for "crab" reported by the FDA-MP, we grouped together blue crab, king crab, and snow crab data. Formal parametric statistical comparisons, such as ANOVA, were not possible for our analyses because the database is composed of aggregate, mean Hg values, as opposed to raw data. Thus, unknown distributions of the underlying Hg data, together with unequal samples sizes for the comparisons of interest, made statistical comparisons inappropriate for our study. Finally, we calculated the percentage of studies reporting a mean Hg concentration exceeding the FDA action level (1 ppm) and the EPA human health criterion (0.3 ppm) for seafood items with relatively higher taxonomic resolution when possible, in order to yield more detailed results than those from broader seafood categories. The FDA action level for methylmercury of 1.0 ppm represents the threshold above which the agency can take legal action (e.g., removing the product from the marketplace) (USFDA 2007). The EPA methylmercury criterion of 0.3 ppm represents the fish tissue concentration that should not be exceeded for safe consumption of sport-caught fish in local waters based on average consumption (USEPA 2001).

To compare farmed items to wild-caught items within the same seafood category, we focused on species with established or emerging, rather than nascent farming or ranching industries. For some seafood categories, the species composition of farmed and wild items is not identical. For example, wild-caught catfish include channel catfish, blue catfish and brown bullhead, while farmed catfish include channel catfish and striped catfish. We designated individual data as farmed or wild according to information from original studies. When farmed or wild status was not reported, as with some market basket studies, we made assumptions based on FAO fisheries statistics for individual species (Food and Agriculture Organization of the United Nations, 2011). Specifically, we assumed lake trout were wild-caught, and rainbow trout

were farmed. For eel species from market studies, we assumed Japanese eel (*Anguilla japonica*) were farmed and European conger eel were wild caught. Finally, we assumed Atlantic salmon from market studies in North America and Europe were farmed unless otherwise specified, given the endangered status of wild Atlantic salmon.

Results

Overview of the Seafood Hg Database

The resulting Seafood Hg Database contains approximately 300 unique data sources (see Supplemental Material, Table S1 (Summary of Hg concentrations across studies in commonly consumed seafood items in the U.S.) and Table S2 (Seafood Hg Database, also available at http://knb.ecoinformatics.org/data.jsp)). In contrast with other, well known compilations of U.S. seafood Hg data (the FDA-MP, the NMFS 1978 report, and combined EPA fish monitoring programs from different regions (e.g. EMAP, REMAP, NCA, etc.), the Seafood Hg Database includes data from both academic and government data sources (approximately 50% of observations from each source type). In addition, the Seafood Hg Database contains large amounts of data on imported fish and shellfish (43% of observations, 21% excluding market studies outside of the U.S. for which exact seafood origin is uncertain).

Variability, Patterns and Information Gaps

We observed relatively high variability in Hg concentrations for individual seafood items. Mean Hg concentrations reported across studies for a given seafood item spanned 0.3 to 2.4 orders of magnitude (for tilefish from the Gulf of Mexico and tuna, fresh/frozen, respectively), mean =1.3 orders of magnitude (Supplemental Material, Table S1). Coefficients of variation for individual seafood items ranged from 0.22 (tilefish from the Gulf of Mexico) to 15.42 (softshell

clams), mean CV = 3.0. We found high variability in Hg content for both broadly defined seafood categories composed of multiple species (e.g., shark, tuna, shrimp), as well as for individual species (e.g. blue crab, *Callinectes sapidus*).

Hg concentrations for wild seafood items were higher than those of farmed items in the same seafood category for all eight seafood categories included in this comparison (Figure 1). Mean Hg concentrations for wild items were 2 to 12-times higher than mean concentrations for farmed counterparts. Both wild and farmed seafood items can have low minimum mean Hg concentrations (for example, minimum mean concentrations of 0.005 and 0.008 for wild and farmed catfish, respectively; Supplemental Material, Table S1). However, wild seafood items generally had higher maximum mean Hg concentrations than farmed seafood items within the same seafood category (for example, maximum mean concentrations of 0.714 and 0.030 for wild and farmed catfish). Finally, we found that, except for Atlantic salmon, farmed seafood items are relatively understudied compared to their wild counterparts based on the total number of samples for each group (Supplemental Material, Table S1).

Our analysis indicated that seafood Hg is understudied in some of the world's most important fisheries. We compared the percentage of studies in the database conducted in major regions in the world (excluding market basket studies) to the percentage of US imports from those regions (National Marine Fisheries Service 2010). Hg in seafood from Asia and South America were understudied, while Hg in seafood from North America (excluding the U.S.) and Europe were well studied, relative to the percent imports from those regions (Figure 2). For example, approximately 60% of seafood imported into the U.S. is from Asia, while only 16% of non-U.S. studies were conducted in Asia. The most studied seafood items, based on the total number of samples measured across studies, include both high Hg items (0.6 to ≥1 ppm) such as

shark (grand mean Hg = 0.882 ppm, 3,722 samples) as well as moderate items (0.3 to 0.59 ppm) such as tuna (0.450 ppm, 3,780 samples) and low Hg items (0 to 0.29 ppm) such as oysters (0.020 ppm, 5,310 samples) (Supplemental Material, Table S1). The least studied items included monkfish (0.174 ppm, 92 samples) and haddock (0.164 ppm, 226 samples) among items with low to moderate Hg, and tilefish (all; 0.883 ppm, 109 samples) and orange roughy (0.513 ppm, 152 samples) among items with moderate to high Hg. We also found few studies on freshwater bass from locations considered important for commercial harvest of these fish (e.g. Great Lakes, Canada). However, there are many studies not included in our framework that report Hg values for bass and other freshwater taxa from locations with recreational fisheries (e.g., Lange et al. 1993).

Comparison with FDA-MP and Federal Criteria

Mean Hg concentrations from the summarized FDA-MP data (FDA Monitoring Program 2011) differed from the grand means estimated from the Seafood Hg Database by 20% or more for more than half of the seafood items in the summarized FDA-MP data (33 out of 58) (Figure 3). Most of these discrepancies were cases in which the FDA-MP estimates for mean Hg content were lower than grand mean estimates from our database (27 out of 33 seafood items). Of these, only marlin, king mackerel, and weakfish/seatrout and freshwater trout were moderate to high Hg seafood items (Figure 3B). In contrast, FDA-MP estimates of mean Hg content were higher than our grand mean for only 6 seafood items, all of which were relatively low in Hg (Figure 3C). Mean values reported for 30 of the seafood items analyzed exceeded the EPA human health criterion of 0.3 ppm in at least 30% of the observations across studies in the database (Figure 4). In comparison, 6 seafood items exceed the FDA criterion of 1 ppm in at least 30% of the observations in our database.

Discussion

Our findings have important implications for estimates of Hg exposure, risk and the development of seafood consumption advice. First, we found discrepancies in mean Hg content estimated by the FDA-MP (2011) compared to the larger Seafood Hg Database, suggesting that consumption advice and exposure estimates based on the FDA-MP data should be revisited. Most of these discrepancies were cases in which the FDA-MP estimates of seafood Hg content were lower than our estimates. The FDA-MP is a market basket study whereas our database contains both market basket studies and research studies in which fish were collected directly from their water source. Thus, FDA-MP estimates may be lower than ours due to differences in methodology. However, FDA-MP sampling methods, and potential mechanisms behind any such bias are unclear. Alternatively, FDA-MP estimates may tend to be lower because estimates based on relatively smaller sample sizes inherently are less likely to include rarer, high values. In general, while the FDA-MP specifically focuses on Hg concentrations in market seafood that are relevant to typical exposures, Hg estimates based on larger sample sizes are inherently more reliable, particularly given the high degree of Hg variability.

Large discrepancies in estimates of seafood Hg content are likely to result in inaccurate estimates of Hg exposure and risk, particularly for high Hg content seafood items and frequently consumed items. For example, marlin (grand mean Hg of 1.517 ppm, 821 samples) currently are not considered high Hg fish according to the FDA-MP (FDA-MP mean Hg of 0.485 ppm, 16 samples), even though marlin have similar Hg concentrations as shark, swordfish, and tilefish from the Gulf of Mexico, for which consumption limits are recommended to reduce risky Hg exposure. The majority of discrepancies, for which FDA estimates of Hg content are lower than our estimates, are for low Hg seafood and are likely to have minor health consequences

compared to discrepancies of moderate to high Hg seafood. However, many of these low Hg seafood items (e.g., shrimp, clams and flounder) are among the most popular with US consumers (Groth 2010). Hence, consumption of these items may result in Hg exposures that exceed previous estimates for the U.S. population. In addition, our results suggest that certain seafood items, such as yellowfin tuna (grand mean Hg of 0.270 ppm, 1183 samples), contain lower Hg concentrations than estimated by the FDA-MP (FDA-MP mean Hg of 0.354 ppm, 231 samples), and that increased consumption of these items may be possible with negligible risk. Our analyses of the percentage of Hg values that exceed federal criteria provide further insight into the seafood items that should be the focus of management and policy development. Finally, we found higher variability in seafood Hg concentrations than previously observed (Sunderland 2007). This high variability reflects the framework of the Seafood Hg Database, which encompasses variability across regions, time, fish size class, and other factors that vary within the overall U.S. market, but are typically constrained within individual studies. Together, the discrepancies and high variability of seafood Hg concentrations we observed based on a large aggregation of data indicate that smaller datasets are more susceptible to random sampling error and may be inadequate aids to develop public health policy or scientific understanding. While smaller, individual datasets may be more accurate for estimating exposures in specific local populations, they may not reflect the full range of seafood Hg concentrations in the U.S. market.

There is a clear need to identify and compare the key sources of variability in seafood Hg content, and translate this information into consumption advice and exposure and risk analyses. Many studies of freshwater fish have identified factors that influence Hg variability. These factors, including physicochemical (pH, dissolved organic carbon, nutrient availability) (Chen et al. 2005; Driscoll et al. 1995) and eco-physiological factors (food chain length, body size)

(Borgmann and Whittle 1991; Cabana et al. 1994; Chen et al. 2000) are often confounded and vary among ecosystems and over time. Compared to the freshwater literature, fewer studies have examined links between Hg content of seafood and factors such as body size and geographic harvest region (Sunderland 2007). Future efforts should account for and identify the key factors influencing Hg content in commercial seafood (e.g., body size, trophic level) as well as compare differences in Hg content among geographic regions. Progress is more likely if large monitoring studies explicitly report data on these factors together with seafood Hg data. Research efforts examining the influence of these factors in commercial fish and shellfish are critical to better predict changes in Hg content of commercial seafood.

Our analyses highlight challenges associated with characterizing variability of seafood Hg across studies and potential sources of bias. Accurate assessments of exposure and risk are ideally derived using probability distributions based on raw data (Sioen et al. 2007; World Health Organization 2000). However, many of the studies that we reviewed, particularly from the academic literature, did not report raw values and less than half of all studies reported standard deviations or standard errors. To capitalize on the abundance of aggregate data in the literature (e.g., mean values), additional studies should test and validate methods used to generate probability distributions (World Health Organization 2000). Our estimates of variability of seafood Hg content are likely to be influenced by the types of available data. For example, differences in data collection methods among studies, such as analysis of fillet versus whole fish, methylmercury versus total Hg values, including samples below detection limits, and differences in fish size (often not reported), each are likely to introduce variability in overall Hg estimates. Moreover, such as geographic and temporal factors, both within and between studies may contribute to our estimates of variability. Standardization or consistent disclosure of

measurement methods would greatly facilitate comparison and aggregation of data into larger datasets that can be used to monitor exposure and risk.

Our results demonstrate that lower Hg concentrations in farmed fish compared to wild fish is broadly consistent, despite high variability typical of fish Hg concentrations across studies, for each seafood item analyzed. However, Hg data for farmed fish are relatively scarce. Thus, there is a need for more extensive study of Hg concentration patterns in farmed compared to wild fish, and the factors that influence them. Nevertheless, given the increase in global consumption of farm-raised fish (National Marine Fisheries Service 2010), their Hg levels should be distinguished from wild fish and explicitly incorporated into consumption advice and risk analyses.

While previous studies have shown lower Hg levels in farmed fish than wild fish, they have typically focused on individual taxa (Balshaw et al. 2008; Dasgupta et al. 2004), primarily salmon (Easton et al. 2002; Foran et al. 2004) and on fish from only a few sources (Dasgupta et al. 2004; Easton et al. 2002). Moreover, the pattern is not universal. At least three studies found no difference in Hg levels between farmed and wild salmon (Easton et al. 2002; Foran et al. 2004) and farmed and wild cod (Jardine et al. 2009). In contrast, our study found consistently lower mean Hg concentrations in farmed seafood across studies for multiple seafood items. In some cases, differences in farmed and wild Hg content may partly reflect taxonomic differences. For example, farmed trout (mostly rainbow trout) have similar Hg concentrations to wild rainbow trout, but lower Hg concentrations compared to wild lake trout. Lower Hg in farmed fish also may be due to ecological characteristics unique to aquaculture settings, such as lower Hg levels in feed, shorter food chain lengths, or a growth dilution effect via higher growth efficiency (Karimi et al. 2010). More broadly, our findings contrast with studies that have found

higher concentrations of persistent organic pollutants (PCBs, dioxins, pesticides) in certain types of farmed fish (Hites et al. 2004; Kelly et al. 2011), possibly reflecting the content of the diet provided in aquaculture operations. Therefore, understanding the mechanisms behind differences in contaminant content in farmed and wild seafood is a necessary step toward effectively managing farmed seafood production.

Our analyses support the need to revise monitoring efforts of both seafood Hg content and characteristics of the U.S. seafood market in order to better track human exposure and potential health risk. In general, monitoring efforts should focus on seafood items that tend to exceed federal criteria (e.g., EPA criterion of 0.3 ppm), that are relatively understudied, or that have highly variable Hg content, in order to better understand seafood Hg concentrations. Specifically, our results suggest a need to increase monitoring of imported seafood from Asia and South America, farmed seafood, and specific seafood items that have been understudied. Increased monitoring efforts may be particularly important for understudied, high Hg seafood items. For example, tilefish is thought to pose a high risk of MeHg exposure (FDA 2004), due to estimates of Hg content for tilefish collected from the Gulf of Mexico in the 1970s (FDA Monitoring Program 2011; Hall et al. 1978). Current estimates of tilefish collected from a more geographically extensive region are needed to test whether tilefish continue to pose a health risk. In addition, improved traceability and transparency of the U.S. seafood market is critical to control Hg exposure and risk by providing information about seafood sources (e.g. country of origin) and taxonomic identity. Complex market linkages, including re-exports of imported fish, change over time and are largely unaccounted for in market data (e.g.,National Marine Fisheries Service 2010), yet are necessary to track exposure from geographic origin of fish to consumers. Growing imports, together with a lack of market traceability (Jacquet and Pauly 2008) and

seafood identification practices (Lowenstein et al. 2009) challenge our ability to estimate exposure, as both geographic origin (Sunderland 2007) and species identity are important determinants of seafood Hg content. Ideal monitoring efforts will need to consider changes in market sources, species composition and size, along with human consumption patterns.

Conclusions

Our findings suggest that seafood consumption advice and exposure estimates based on smaller datasets such as the FDA-MP should be revisited using larger datasets that are more likely to capture accurate estimates of mean Hg values and their variability in U.S. commercial seafood. Priorities for new research should include increased monitoring of farmed seafood and imported seafood from Asia and South America, as well as studies examining the processes underlying lower Hg concentrations in farmed seafood. Finally, additional studies should compare the relative influence of different environmental and ecological factors on the variability of seafood Hg content.

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Figure Legends

Figure 1. Grand mean Hg ± SE in farmed (dark shaded bars) and wild seafood items (light shaded bars).

Figure 2. Percentage of all imports into the U.S. by volume in 2009 according to region (National Marine Fisheries Service 2010) (dark shaded bars) and percentage of all non-US studies in the Seafood Hg Database by region conducted in foreign countries (light shaded bars). Studies from foreign countries exclude market basket studies. Percent imports do not account for re-exportation of imported fish.

Figure 3. Mean Hg content estimated from the Seafood Hg Database and the FDA-MP for 58 seafood items, compared to the 1:1 line. Inset, mean Hg content estimates compared to the 1:1 line for seafood items with mean Hg ≤ 0.3 ppm. **(A)**. Seafood items for which FDA-MP underestimates of mean Hg content are lower than mean estimates based on the Seafood Hg Database (Discrepancy = Hg Database Mean / FDA-MP Mean) **(B)**. Seafood items for which FDA-MP overestimates of mean Hg content are higher than the Seafood Hg Database (where Discrepancy = FDA-MP Mean / Hg Database Mean, or the inverse of Discrepancy in Figure 3B) **(C)**. Larger discrepancy values >1 indicate larger difference. Seafood items for which discrepancy <20% are excluded from figure.

Figure 4. Percent of studies reporting mean Hg concentrations exceeding federal criteria (FDA, 1 ppm; EPA, 0.3 ppm) per seafood item, shown with increasing % exceedances of EPA criterion from left to right. Figure excludes taxa with <30% exceedances.

Figure 1.

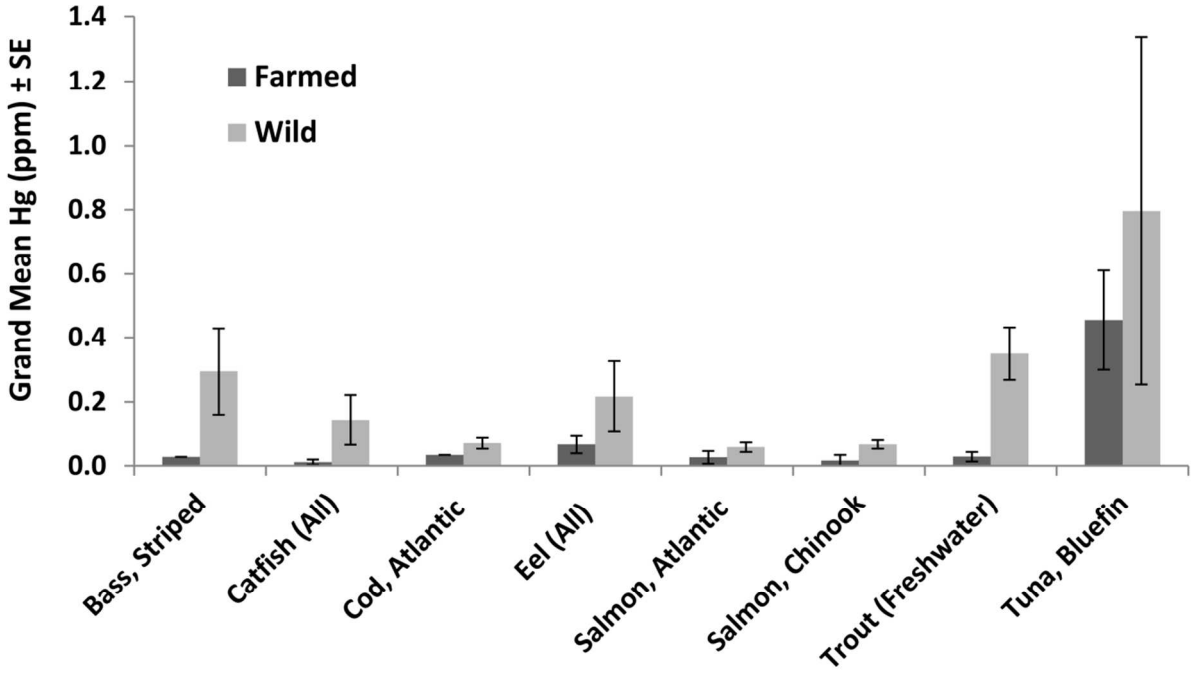

Grand Mean Hg (ppm) ± SE
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
Farmed
Wild
Bass, Striped
Catfish (All)
Cod, Atlantic
Eel (All)
Salmon, Atlantic
Salmon, Chinook
Trout (Freshwater)
Tuna, Bluefin

Figure 2.

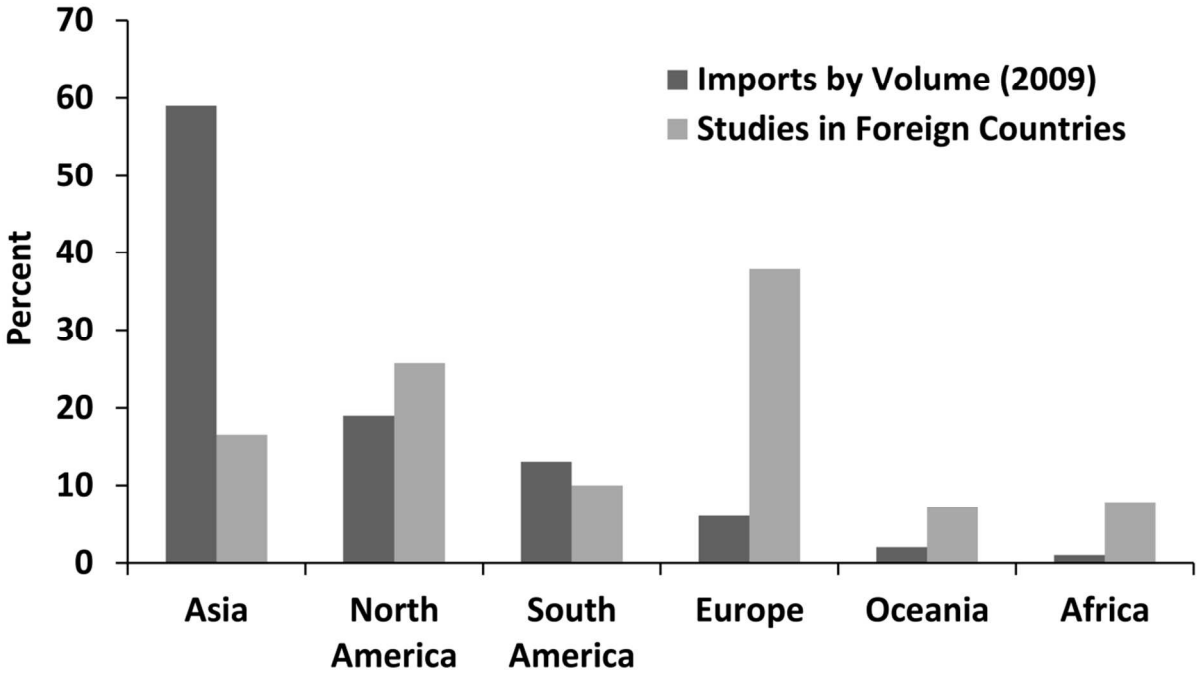

70
60
50
40
30
20
10
0
Percent
Imports by Volume (2009)
Studies in Foreign Countries
Asia
North America
South America
Europe
Oceania
Africa

Figure 3

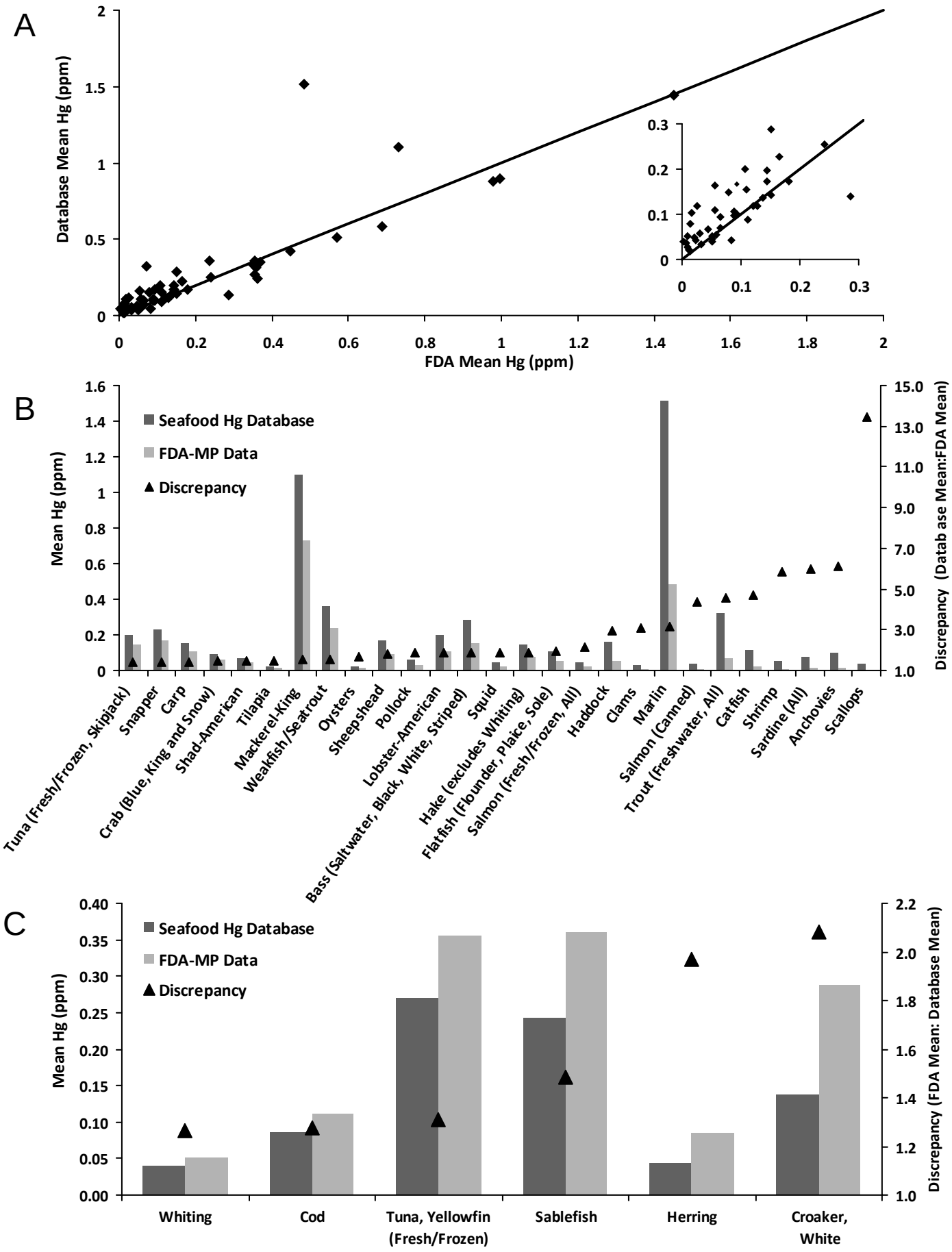
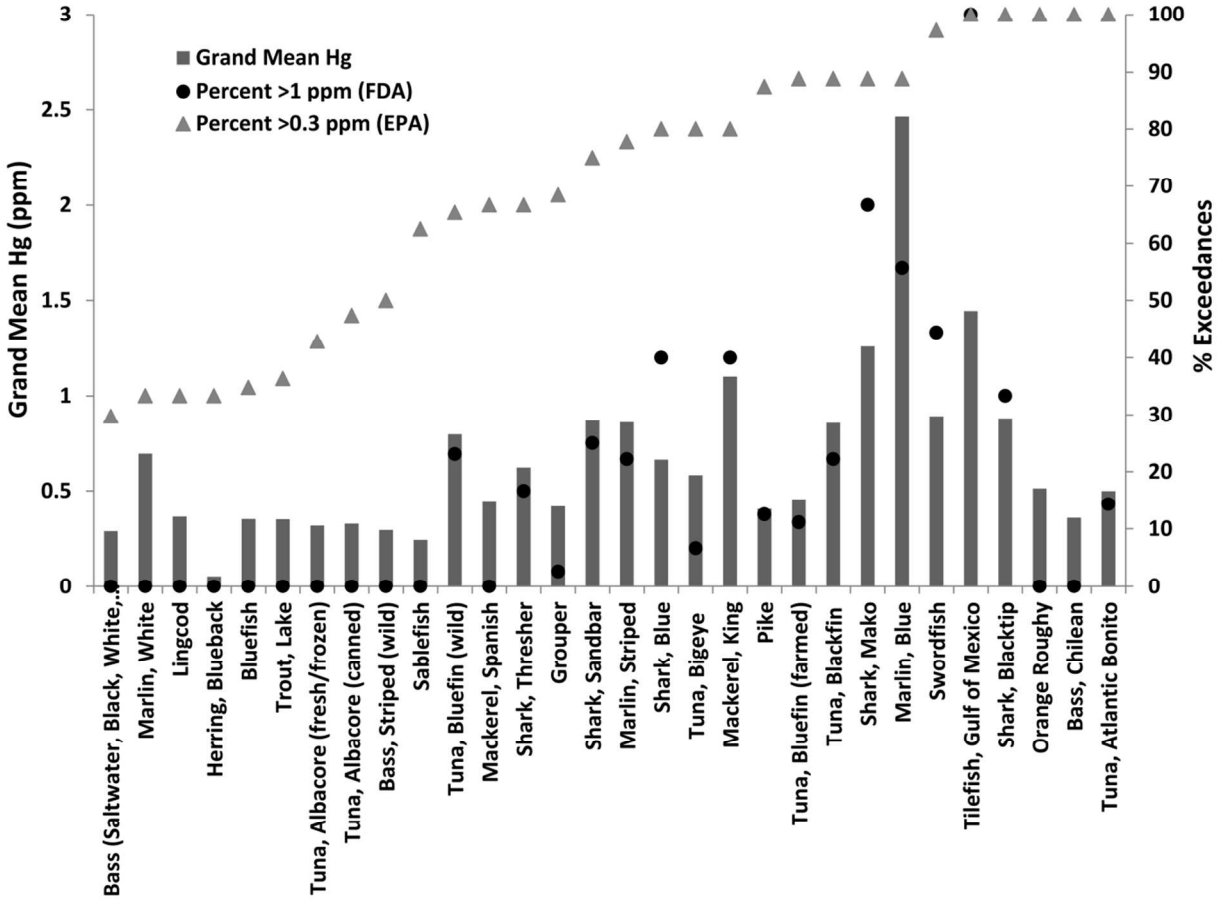

A
Database Mean Hg (ppm)
FDA Mean Hg (ppm)
B
Seafood Hg Database
FDA-MP Data
Discrepancy
Mean Hg (ppm)
Discrepancy (Datab ase Mean:FDA Mean)
Tuna (Fresh/Frozen, Skipjack)
Snapper
Carp
Crab (Blue, King and Snow)
Shad-American
Tilapia
Mackerel-King
Weakfish/Seatrout
Oysters
Sheepshead
Pollock
Lobster-American
Bass (Saltwater, Black, White, Striped)
Squid
Hake (excludes Whiting)
Flatfish (Flounder, Plaice, Sole)
Salmon (Fresh/Frozen, All)
Haddock
Clams
Marlin
Salmon (Canned)
Trout (Freshwater, All)
Catfish
Shrimp
Sardine (All)
Anchovies
Scallops
C
Seafood Hg Database
FDA-MP Data
Discrepancy
Mean Hg (ppm)
Discrepancy (FDA Mean: Database Mean)
Whiting
Cod
Tuna, Yellowfin (Fresh/Frozen)
Sablefish
Herring
Croaker, White

Figure 4.


Grand Mean Hg
Percent >1 ppm (FDA)
Percent >0.3 ppm (EPA)
Grand Mean Hg (ppm)
% Exceedances
3
2.5
2
1.5
1
0.5
0
100
90
80
70
60
50
40
30
20
10
0
Bass (Saltwater, Black, White,..
Marlin, White
Lingcod
Herring, Blueback
Bluefish
Trout, Lake
Tuna, Albacore (fresh/frozen)
Tuna, Albacore (canned)
Bass, Striped (wild)
Sablefish
Tuna, Bluefin (wild)
Mackerel, Spanish
Shark, Thresher
Grouper
Shark, Sandbar
Marlin, Striped
Shark, Blue
Tuna, Bigeye
Mackerel, King
Pike
Tuna, Bluefin (farmed)
Tuna, Blackfin
Shark, Mako
Marlin, Blue
Swordfish
Tilefish, Gulf of Mexico
Shark, Blacktip
Orange Roughy
Bass, Chilean
Tuna, Atlantic Bonito